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Singlet-triplet minus mixing and relaxation lifetimes in a double donor dot

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We measure singlet-triplet mixing in a precision fabricated double donor dot comprising 2 and 1 phosphorus atoms separated by 16 ± 1 nm. We identify singlet and triplet-minus states by performing a sequential independent spin readout of the two electron system and probe its dependence on magnetic field strength. The relaxation of singlet and triplet states is measured to be 12.4 ± 1.0 s and 22.1 ± 1.0 s, respectively, at $B_z = 2.5$ T. *Published by AIP Publishing.*
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Single electrons confined to donor atoms in silicon are ideal candidates for semiconductor spin qubits due to their long spin relaxation[1,2] and coherence lifetimes.[3,4] In these systems, the coupling of two electrons offers not only the ability to perform quantum logic between multiple qubits[5,6] but also the potential for different qubit types.[7] For example, singlet-triplet-zero qubits are appealing as they are immune to fluctuations in local magnetic field strength[8] and singlet-triplet-minus qubits can be operated via electrical gates alone in the presence of a sufficiently large inhomogeneous magnetic field.[9]

In donor based devices, the operation of multi-electron systems relies heavily on the ability to engineer a precise value of coupling between them.[10] In cases where exchange coupling is desired, due to its exponential dependence on the inter-donor distance, the placement of donors inside the silicon lattice must be done with close to atomic precision.[11,12] This can be achieved using scanning tunnelling microscopy (STM) hydrogen resist lithography, which has previously shown this level of donor placement.[13]

While the relaxation of single electrons confined to donors has been well understood for over 40 years,[14–17] there has been far less attention to two-electron spin relaxation processes[18,19] since most of the two-donor papers focus on calculating the exchange energy, J.[20,21] Theoretical calculations of two-electron relaxation in Ref. 18 predict a J^3 dependence on the spin relaxation rate, whereas the inclusion of spin-orbit coupling predicts a J^4 or J^5 depending on the temperature.[19] The results from gate-defined quantum dots also predict a dependence of the singlet-triplet relaxation[22] on the exchange (or detuning)[23,24] and magnetic field.[23,25] Previous experimental studies have measured singlet-triplet relaxation rates of strongly coupled electrons confined to donors, Ref. 26 (exchange energy of $J \approx 300$ μeV) and Ref. 27 (tunnel coupling $t_c = 47$ μeV), finding triplet-state relaxation rates of the order 100 ms and 60 ns, respectively. In the latter work, an STM fabricated double donor dot was probed in transport[27] where the interaction of the source and drain leads with the double dot itself led to the observed fast spin relaxation of the singlet-triplet states.

In this paper, we use a charge sensed double donor dot fabricated with precision placement by means of STM lithography to probe singlet-triplet relaxation of a two-electron state at the (1,1)–(2,0) inter-dot charge transition. The two dots L and R comprise 2 and 1 P donors, respectively. The details of device fabrication and characterisation have been reported previously in Ref. 28. Figure 1(a) shows a close up STM image of the device which comprises the two donor dots both of which are tunnel coupled to a larger quantum dot of approximately 1000 P atoms. This larger dot is itself tunnel coupled to source (S) and drain (D) leads, across which we apply a bias of $V_{SD} = 2.5$ mV such that it operates as a single electron transistor charge sensor. Four phosphorus doped gates $\{G_L, G_M, G_R, G_S\}$ surround the

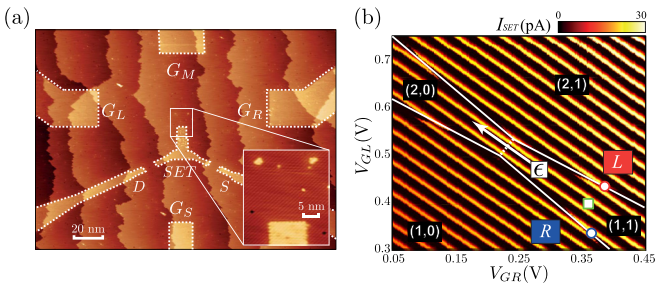

FIG. 1. Precision fabricated donor double dot device. (a) A scanning tunnelling micrograph of the device. Two donor dots L and R separated by 16.0 ± 1.0 nm are composed of 2 and 1 P donors, respectively. Surrounding the donor dots are three gates G_L, G_M, and G_R used to control their electrostatic environment. Approximately 20 nm from the dots is a single-electron-transistor (SET) which itself is a larger quantum dot tunnel coupled to source and drain leads. The SET also acts as an electron reservoir for L and R and is controlled by the gate G_S. The inset shows a close up of the dots and the top of the SET island. (b) A charge stability map of the device shown by the transport current through the SET as a function of the gates $\{G_L, G_R\}$ near the inter-dot charge transition (1,1)-(2,0). Current peaks running at ~45° show the Coulomb blockade of the SET, and breaks in these correspond to transitions of single electrons from either dots L or R, indicated by the solid white lines. The detuning axis ϵ is given by the white arrow (with a lever arm along the axis, $\alpha_\epsilon = 0.071$ eV/V), and the approximate spin-readout positions for L and R are shown by the red and blue circles, respectively. The lever arms of the dots along the spin readout axis (same as the detuning axis) are $\alpha_L = 0.041$ eV/V and $\alpha_R = 0.030$ eV/V. Only one other charge transition corresponding to dot-L is seen at much lower gate voltages, leading to the assignment of the charge regions shown here.

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device and are used for electrical control. Figure 1(b) shows the Coulomb blockade through the charge sensing single-electron-transistor (SET) shown as a function of gate voltages V_{GL} and V_{GR}. Breaks in the current peaks represent charge transitions from either dot-*L* or -*R*, which are used for the spin-readout of these dots at the positions marked by the red and blue circles in Fig. 1(b), respectively. The device was measured in a dilution refrigerator with a base temperature of 100 mK and an electron temperature of ∼200 mK.[1]

We perform single-shot spin readout of the single electrons confined to dots-*L* and -*R* at the 1 → 2 and 0 → 1 transitions, the so-called D^- and D^0 readout schemes, respectively[2] [see Fig. 1(b)]. Both readout schemes rely on an energy selective tunnelling to or from the donor dot to provide a readout signal from the charge sensing SET. The D^0 method (employed for dot-*R*) relies on the Zeeman split spin-up electron tunnelling to the SET following which a spin-down electron tunnels back onto the dot. Conversely, the D^- scheme (for dot-*L*) relies on the fact that the chemical potentials of spin-up and -down electrons transitioning to the two-electron singlet state are also split by the Zeeman energy, allowing for an equivalent readout signal.[2] The advantage of utilising these two readout schemes in conjunction is that we do not need to pulse to the equivalent 0 → 1 transition for dot-*L* which is over 200 mV away from the (1,1)–(2,0) inter-dot charge transition.

The eigenspectrum of two exchange coupled electrons in a magnetic field is shown in Fig. 2(a). The spin-1 triplet-states $|T^+\rangle$ and $|T^-\rangle$ are split from $|T^0\rangle$ by the Zeeman $g\mu_0 B_z$, crossing the singlet states at separate detuning points where mixing between them can occur. This process necessarily involves a spin-flip between one electron spin and a surrounding nuclear spin driven by the hyperfine interaction. Since the P nuclear spins at sites *L* and *R* are limited in our case to ≤2 each, an equal number of electron-nuclear spin flips can occur within the T_1 times of the P nuclei (∼minutes for natural silicon[29]). Therefore, the process driving $|T^-\rangle \leftrightarrow |S\rangle$ mixing will most likely be dominated by a constantly fluctuating ^{29}Si Overhauser field[30] near the $|T^-\rangle \leftrightarrow |S\rangle$ anti-crossing. It has been recently predicted that the spin-orbit coupling could play a significant role in the two-electron relaxation processes of donors for large exchange;[19] however, for the tunnel coupling in this device, which was estimated to be ∼200 MHz, the hyperfine driven relaxation should still dominate the relaxation process.[31]

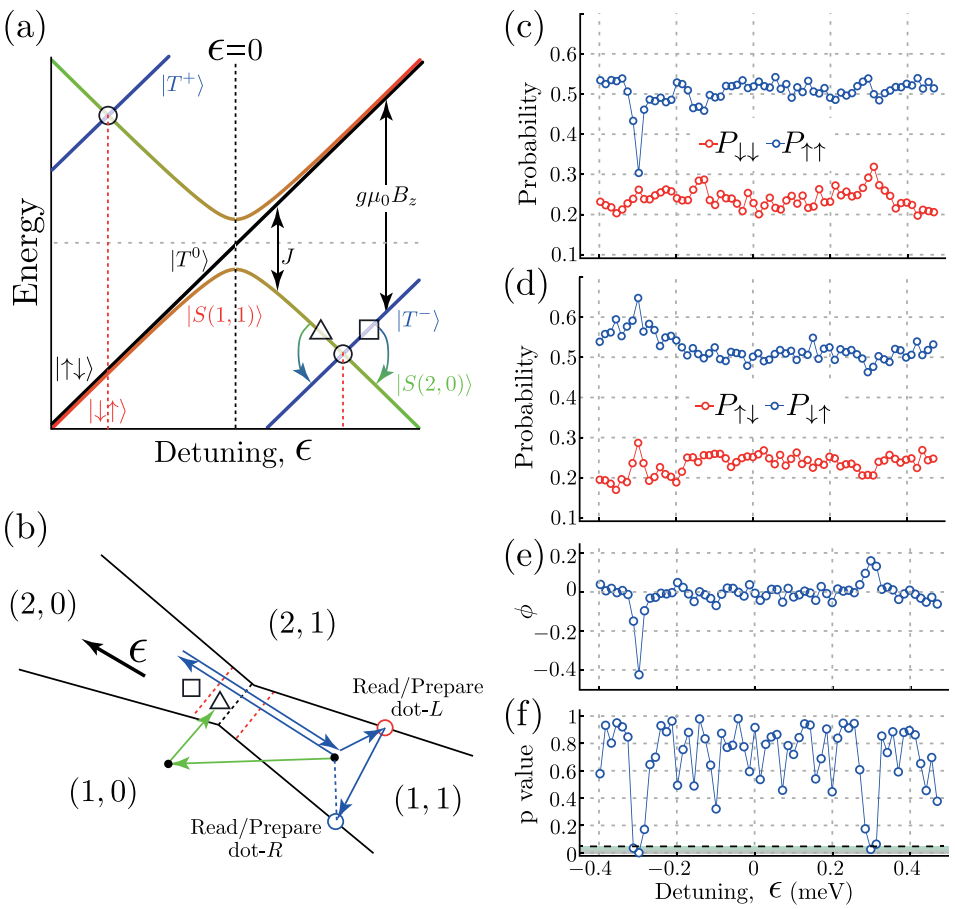

FIG. 2. Observation of mixing between $|S(2,0)\rangle$ and $\{|T^+\rangle, |T^-\rangle\}$ states. (a) The eigenspectrum of the two-electron Hamiltonian showing the dependence of the singlet-triplet states as a function of detuning. Mixing between $\{|T^+\rangle, |T^-\rangle\} \leftrightarrow |S(2,0)\rangle$ occurs at the positions marked by white circles. (b) The pulse sequence used to perform the two different relaxation measurements. The green line shows the pulse to load the $|S(2,0)\rangle$ state via the (1,0) charge region and measure the relaxation of $|S(2,0)\rangle \rightarrow |T^-\rangle$ at the triangle. The blue line shows the pulse to measure the relaxation of $|T^-\rangle \rightarrow |S(2,0)\rangle$ at the square position in detuning. (c) and (d) To further investigate the mixing between the singlet and triplet states, we prepare the fully mixed state ρ_I and apply a 50 ms pulse along ϵ after which we measure the probabilities (c): $P_{\downarrow\downarrow}$ and $P_{\uparrow\uparrow}$ (offset by +0.25 for clarity) and (d): $P_{\uparrow\downarrow}$ and $P_{\downarrow\uparrow}$ (offset by +0.25). (e) The correlation coefficient ϕ as a function of detuning for the data in (c) and (d). For $\epsilon \sim -0.3$ meV, a decrease in ϕ indicates the mixing between $|T^+\rangle \leftrightarrow |S(2,0)\rangle$, whereas an increase in ϕ at $\epsilon \sim 0.3$ meV is where $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ occurs. (f) The p-value of a χ^2 test as a function of ϵ, showing statistically significant mixing of the electron spins; $p \ll 0.05$ (green shaded region) at $\epsilon = \pm 0.3$ meV.

To see this mixing process, we start by preparing a random mixture of spin-up and -down on both L and R, such that we have equal populations of all four two-electron product states,

$$\rho_I = \frac{|\uparrow\uparrow\rangle\langle\uparrow\uparrow| + |\uparrow\downarrow\rangle\langle\uparrow\downarrow| + |\downarrow\uparrow\rangle\langle\downarrow\uparrow| + |\downarrow\downarrow\rangle\langle\downarrow\downarrow|}{4}. \quad (1)$$

A 50 ms pulse is applied from the midpoint between the two readout positions along the detuning axis ϵ towards the (1,1)–(2,0) charge degeneracy. Following this, we return to the readout positions and perform a single-shot spin-readout at L and R in that order, and a schematic representation of the full pulsing sequence is shown in Fig. 2(b). The sequence is repeated 100 times for different values of ϵ, and the joint spin-readout probabilities P_{ij} are shown in Figs. 2(c) and 2(d). The probability of $P_{\uparrow\uparrow}$ in Fig. 2(c) shows a sharp decrease at approximately -0.3 meV, whereas both $P_{\uparrow\downarrow}$ and $P_{\downarrow\uparrow}$ show an increase at this value of detuning, indicating mixing between $|T^+\rangle \leftrightarrow |S(2,0)\rangle$ [see Fig. 2(a)]. The opposite effect is observed for mixing between $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ at approximately 0.3 meV. The position of the two singlet-triplet mixing points is used to calibrate the zero detuning point $\epsilon = 0$.

These results can be understood by considering the relaxation processes occurring in the system. Any increase in $|S(2,0)\rangle$ due to mixing between $|T^+\rangle \leftrightarrow |S(2,0)\rangle$ at negative detuning [white circle at $\epsilon < 0$ in Fig. 2(a)] will result in a fast charge relaxation from $|S(2,0)\rangle \rightarrow |S(1,1)\rangle$, leading to a *decrease* in the overall $|T^+\rangle$ population after $|T^+\rangle \leftrightarrow |S(2,0)\rangle$ mixing. However, at positive detuning, relaxation from the $|S(1,1)\rangle \rightarrow |S(2,0)\rangle$ (and $|T^0\rangle \rightarrow |S(2,0)\rangle$) will result in an *increase* in $|T^-\rangle$ due to mixing between $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ [white circle at $\epsilon > 0$ in Fig. 2(a)]. The resulting correlation coefficient ϕ and its statistical significance are given in Figs. 2(e) and 2(f), respectively.[31] A significantly negative value of ϕ for $\epsilon < 0$ and a positive value for $\epsilon > 0$ follow as a result of the mixing described above; furthermore, its values at these positions are statistically significant with a χ^2 p-value of $\ll 0.05$ in both cases.

Providing further evidence that the above results are consistent with the $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ mixing, we repeat the equivalent pulsing scheme with the initial state $\rho_i = |\downarrow\downarrow\rangle\langle\downarrow\downarrow|$ for different values of static magnetic field, B_z. This state can be prepared deterministically, as the spin readout process leaves a spin-down electron. The results of this experiment are shown in Fig. 3. We see that from Fig. 2(a) that the position of singlet-triplet minus mixing occurs where the exchange energy is equal to the Zeeman splitting, that is, where $J = g\mu_0 B_z$, which can clearly be seen in our data. Importantly, for devices where the tunnel coupling t_c is of the order of the Zeeman, a non-linear dependence of singlet-triplet mixing should be observed as in the so-called *spin-funnel* experiments.[32] However, the measurements shown in Fig. 3 are restricted to $B_z \geq 2.5$ T in order that a high fidelity spin readout can be performed, and the position of $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ mixing therefore shows a linear dependence on B_z, placing a bound on the tunnel coupling $t_c \ll 2.5 g\mu_0$.

In this last section, we present the results of singlet-triplet relaxation. The relaxation of singlet-triplet states can

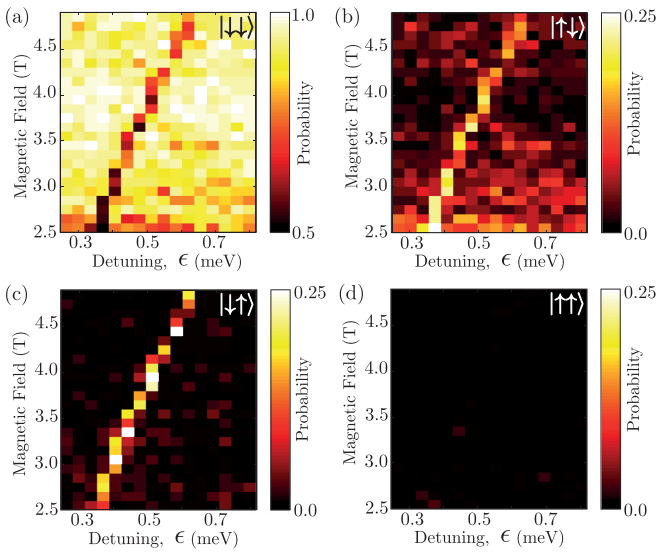

FIG. 3. Mixing between $|S(2,0)\rangle$ and $|T^-\rangle$ as a function of the magnetic field and detuning. The initial state $|T^-\rangle = |\downarrow\downarrow\rangle$ is prepared by means of reading both the left and right dot electrons, after which a 50 ms pulse is applied along the detuning axis ϵ. Upon returning to the readout positions, the probabilities $\{P_{\downarrow\downarrow}, P_{\uparrow\downarrow}, P_{\downarrow\uparrow}, P_{\uparrow\uparrow}\}$ are measured. Mixing between $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ is apparent where a decrease in $P_{\downarrow\downarrow}$ is observed, shown in (a), at the expense of an increase in the anti-correlated probabilities $P_{\uparrow\downarrow}$ and $P_{\downarrow\uparrow}$ shown in (b) and (c), respectively. (d) The probability $P_{\uparrow\uparrow}$ is not affected and therefore shows no significant change. The magnitude of the exchange energy, $J = \gamma_e B_z$, can be seen to depend linearly on the corresponding detuning value, ϵ.

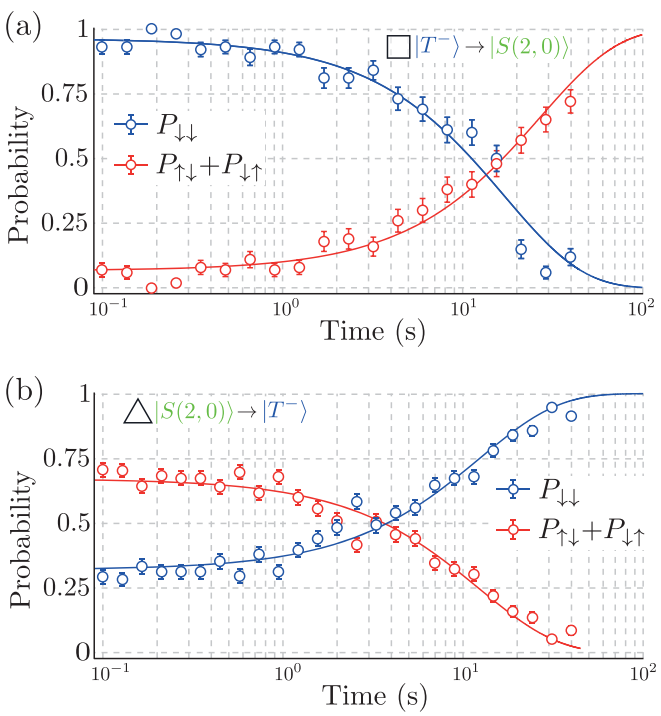

FIG. 4. Relaxation of singlet and triplet states as measured by a two-electron spin readout. (a) The relaxation of $|T^-\rangle$ into $|S(2,0)\rangle$ measured at $B_z = 2.5$ T. The probability of measuring anti-correlated electron spins $P_{\uparrow\downarrow}$ and $P_{\downarrow\uparrow}$ increases as a function of wait time, indicating relaxation into the singlet state. A fit to the data gives a relaxation time, $T_1 = 22.1 \pm 1.0$ s. (b) Relaxation of $|S(2,0)\rangle$ into $|T^-\rangle$ can be seen by an increasing $P_{\downarrow\downarrow}$ probability with wait time. A fit to the data gives a relaxation time, $T_1 = 12.4 \pm 1.0$ s.

both be probed around the $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ mixing point [see Fig. 4(a)]. After deterministically loading $|T^-\rangle$, we pulse to a position 1 mV positively detuned from the $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ mixing point where we wait up to 40 s [see Fig. 4(a)]. Here, $|S(2,0)\rangle$ is the ground state, and we see that the probabilities $P_{\uparrow\downarrow}$ and $P_{\downarrow\uparrow}$ increase as $P_{\downarrow\downarrow}$ decreases as a result of relaxation with $T_1 = 22.1 \pm 1.0$ s. To deterministically prepare the singlet state, $|S(2,0)\rangle$, we perform the pulse sequence shown by the green arrows in Fig. 2(b). After unloading an electron from R by pulsing into the (1,0) charge region, we pulse to the (2,0) charge region 1 mV negatively detuned from the $|T^-\rangle \leftrightarrow |S(2,0)\rangle$ mixing point. In the (2,0) region, only a singlet state can be loaded due to the Pauli spin blockade.[27,33] At this position, $|T^-\rangle$ is the ground state, and relaxation of the singlet state occurs with $T_1 = 12.4 \pm 1.0$ s [see Fig. 4(b)]. Note that for this experimental run, the spin readout fidelity of the left-dot was significantly reduced due to misalignment of the spin readout voltage level, and this accounts for the increased value of $P_{\downarrow\downarrow}$ from $t = 0$, that is, we were more likely to count $|\uparrow\rangle$ as $|\downarrow\rangle$. The value of T_1 measured here is in stark contrast to a previously measured relaxation time of 60 ns,[27] where coupling to in plane leads meant that the two-electron state was poorly isolated, causing rapid relaxation of the singlet state via spin exchange with the leads.

The coupling of multiple electron spins is a prerequisite for scalable solid-state quantum computation. The resulting entangled states can be utilised directly by performing a single-shot spin readout[5] or even as a qubit subspace.[9,32] The long T_1 lifetimes of singlet and triplet states measured here do not present as a challenge for future experiments and were expected given previous results on single spin states of donors.[1,2,4] However, the coherent characteristics of such states are unknown, and investigations of them will require either the direct manipulation in a singlet-triplet qubit subspace[9] or similar protocols for the individual electron spins.[5]

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